



LINION CARBIDE CORPORATION

NUCLEAR DIVISION

P. O. BOX Y, OAK RIDGE, TENNESSEE 37830

January 17, 1974

United States Atomic Energy Commission Attention: Mr. William H. Travis, Director Safety & Environmental Control Division Post Office Box E Oak Ridge, Tennessee 37830

Gentlemen:

Inventory of Radioactivity Released to Onsite and Offsite Environments

The attached information is submitted in response to your letter of October 18, 1973. The data tabulated represent the best information available on releases of radioactivity to onsite and offsite environments.

In general, data have been tabulated on a yearly basis and no decay corrections have been applied. Information previously reported via other systems has been referenced only. The completeness of the information is limited by the unavailability of old records. In some cases, quantitative data simply are nonexistent.

We will be happy to discuss the data with you or members of your staff.

Very truly yours,

R. G. Jordan, Manager

Safety and Environmental Protection

RGJ:ayb

Attachment

cc: Mr. P. C. Fourney

Mr. R. F. Hibbs

Dr. H. Postma

Mr. P. R. Vanstrum - 6

APPROVAL FOR RELEASE

Unnumbered report dtd 1/17/74, TNVENTORY OF RADIOACTIVITY RELEASED TO ONSITE AND

Title/Subject OFFSITE ENVIRONMENTS; and 1-page 1tr,

RG Jordan to WH Travis (AEC-OR), 1/17/74

Approval for unrestricted release of this document is authorized by the Oak

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K-25 Classification & Information Control Officer

Date

INVENTORY OF RADIOACTIVITY RELEASED TO ONSITE AND OFFSITE ENVIRONMENTS

Union Carbide Corporation - Nuclear Division

January 17, 1974

The attached information presents a tabulation obtained from currently available records of the radioactivity released to onsite and offsite environments of the UCC-ND-operated installations.

Oak Ridge National Laboratory

Tables 1 through 8 present an inventory of the quantities and locations of radioactivity released to the environment from ORNL. Information regarding the amount of radioactivity deposited in the burial grounds and into the soil resulting from spills can be found in the ORNL Radioactive Waste Management Plans, ORO-723 (ORNL), issued June 25, 1973. ORO-723 also gives estimates of the activity in the Process Waste Settling Basin; however, no estimate is given for the amount of activity contained in the sediment of White Oak Lake. A comprehensive study of White Oak Lake sediment in December 1962 indicated that, at that time, approximately 1038 curies of 106Ru, 704 curies of 237Cs, 152 curies of 60Co, 17 curies of trivalent rare earths, and 15 curies of 90Sr were contained in the sediment.

Reliable data regarding gaseous discharges prior to the installation of the instack sampler at the 50-ft level of the 3039 stack are not available. The instack sampler was installed in the Fall of 1961. Curie quantities of the specific radionuclides in the waste solutions pumped to the waste pits and trenches are not available prior to 1961. Prior to 1961, the waste content was reported in terms of gross beta activity only. The amount of ²³⁹Pu deposited in these trenches and pits is 634 grams as reported in ORO-723.

Tables 1, 2, 3, and 4 present the annual discharges of waste to Pits 2, 3, and 4; Trench 5; Trench 7A and Trench 7B, respectively. Table 5 presents the annual discharge of radionuclides to the Clinch River. Table 6 presents the annual deposition of ⁹⁰Sr in the sludge removed from the Waste Treatment Plant. Table 8 gives the annual discharge of radionuclides to the atmosphere beginning in 1962. Unless stated otherwise, the values shown in the tables were obtained from analyses and associated flow or volume data. No decay corrections have been applied to the quantities tabulated.

Paducah Gaseous Diffusion Plant

Tables 9 through 11 present an inventory of the quantities and locations of radioactivity released to the environment from the Paducah Gaseous Diffusion Plant. Many of the quantities were determined by estimates and extrapolations. The bases for the estimated quantities are given as footnotes at the bottom of each table. Table 9 presents estimated annual releases of uranium to the atmosphere and streams offsite and to the soil onsite from 1952 through 1972. Table 10 gives the uranium discards to the holding pond and burial areas onsite. Table 11 presents an estimate of the ²³⁰Th released from 1956 through 1972.

Trace quantities of neptunium and plutonium were present in ${\rm UO}_3$ reactor tails which have been received at Paducah from Hanford and Savannah River for processing over the past 20 years. Although the quantity per ton was quite small, the total quantity received over the period of years is of possible concern.

A plant inventory and material balance were made of neptunium and plutonium using the estimated receipts and data from limited sampling of various parts of the process system and materials on hand. The study indicated that about 270 grams Pu were received at Paducah from about July 1953 through November 1973. Approximately 13,500 grams Np were received from December 1956 through November 1973. From available data, material balances indicated that approximately 4450 grams Np and 240 grams Pu are unaccounted for.

Considering the process chemistry of the transuranic elements, it appears possible that a significant fraction of this unaccounted for material could still be onsite. A study could be initiated to attempt to evaluate the quantity of these materials that may still remain in the plant. However, this study would be limited by the availability of old records, for example, UF₆ cylinders filled and later washed or fluorination ash processed, and the fact that very few transuranic analytical measurements were made on most materials during this 20-year period. It is likely that only estimated values using very substantial uncertainty can be developed for Np and Pu discharged or still on hand.

In addition to the Np and Pu, traces of ^{99}Tc were also present in the UO $_3$ received from Hanford and Savannah River. From material balances, it is estimated that during this 20-year period about 1200 curies of ^{99}Tc were unaccounted for. A significant portion of this may have been released to the atmosphere and surface streams.

Oak Ridge Y-12 Plant

Table 12 presents an inventory of the quantities of uranium and thorium released from the Y-12 Plant. The curie quantities of uranium released were calculated from data contained in the Y-12 Nuclear Material Inventory Reports using the appropriate enrichment factor for the materials released each year. Information regarding the amount of material deposited in burial grounds and in the S-3 ponds prior to FY 1972 has been reported through the NMIS System. Uranium deposited in the S-3 ponds for FY 1972 was 1.6 curies.

Oak Ridge Gaseous Diffusion Plant

The quantities of material released from the ORGDP for the period 1946 through 1972 are shown in Table 13. This table is a summary of the available information found in release reports, discard records, effluent sampling data, and analytical information.

Table 14 shows five locations at which material from the ORGDP has been buried onsite or offsite:

K-1407-B Holding Pond and ORGDP Burial Ground. The quantity shown is the result of the summation of "discard records" for the period 1959 through 1972.

ORNL and Y-12 Burial Grounds. The quantity of material is cumulative from 1946 through 1964.

Sludge Disposal Area. The material is sludge which was removed from the K-1407-B holding pond in 1961.

Sampling of the sediment in the bed of Poplar Creek was performed during 1959. An estimated 4.8 curies of uranium were in the creek bed at that time.

Reactor tails from Hanford and Savannah River also were processed at the ORGDP during the operation of the feed manufacturing facility. The quantity processed, however, was only about 15% as large as the quantity processed at Paducah. Assuming the ORGDP processed material contained the same concentrations of transuranics as the Paducah material, the transuranic situation would be similar to that discussed previously for the Paducah Plant, but to a much lesser degree.

TABLE 1
Oak Ridge National Laboratory

Radioactive Waste (In Curies) Transferred to Pits 2, 3, and 4

Date	90 _{Sr}	137 _{Cs}	106 _{Ru}	TRE	Estimated Gross Beta
1951	(Test Pit No. 1)				390
1952	•				953
1953					7,716
1954					7,224
1955					21,391
1956					34,989
1957					41,918
1958					52,795
1959					280,000
1960					21,494
1961	92	12,889	757	837	

TABLE 2

Oak Ridge National Laboratory

Radioactive Waste (In Curies) Transferred to Trench 5)

Date	90 _{Sr}	37 _{Cs} 60 ₁		⁶⁰ Co	TRE	Estimated Gross Beta		
1960			-			3,536		
1961	1,116	13,121	830		41			
1962	1,354	14,749	1,274	153	608	•		
1963	4,851	48,108	1,096	837				
1964	5,303	58,688	118	110				
1965	80,243	57,435	388	1,827				
1966	2,719	7,471	24	81				

TABLE 3

Oak Ridge National Laboratory

Radioactive Waste (In Curies) Transferred to Trench 7-A

Date	⁹⁰ Sr	137 _{Cs}	106 _{Ru}	60 _{Co}	TRE
1962	38	1,588	358	11	6
1963	5,250	26,296	1,130	365	
1964	10,583	53,398	189	131	
1965	5,534	28,451	53	125	
1966	3,010	4,576	. 15	136	

TABLE 4

Oak Ridge National Laboratory

Radioactive Waste (In Curies) Transferred to Trench 7-B

Date	90 _{Sr}	137 _{Cs}	106 _{Ru}	⁶⁰ Co	TRE
1962	32	1,668	307	9	5
1963	6,628	25,947	981	385	
1964	6,867	35,876	126	88	
1965	7,327	34,088	_. 54	158	
1966	2,602	4,339	12	12	

TABLE 5

Oak Ridge National Laboratory

Radioactive Waste (In Curies) Disposed of by Hydrofracture

Date	90 _{Sr}	137 _{Cs}	106 _{Ru}	60 _{Co}	239 Pu(gms)	244 Cm(gms)
1964	610	317	36	4		
1965	822	4,920	4	15		
1966	3	19,950	21 .	8		
1967	10,050	75,500	594	642		
1968	4,800	121,300	500	100	36	
1969	8,900	89,000	100	200	4	
1970	2,747	44,833	236	72	29	
1972	3,024	93,135	3,819	157	· 13	2

TABLE 6

Oak Ridge National Laboratory Yearly Discharges of Radionuclides to Clinch River (Curies) $^{\rm a}$

Transuranic Alpha Emitters													0.0004	0.01	0.001	0.07	90.0	0.2	0.08	0.5	0.2	-	0.04	0.2	7.0	0.05	0.05
3 _H																			1929	1161	3090	13273	9685	12247	9473	8945	10600
OD 09										9.9	46	4.8	8.7	77	72	31	14	14	15	12	7	ო	ᅮ			0.8	1.25
1311				77	19	18	20	2.1	3.5	7.0	3.5	1.2	8.2	0.5	5.3	3.7	0.36	0.44	0.29	0.20	0.24	0.91	0.31	0.54	0.32	0.21	0.34
95Nb				22	42	2.2	18	3.6	9.5	5.7	15	7.1	0.9	30	45	70	7.7	0.71	0.07	0.33	0.67	0.49	0.27	0.18	0.02	0.01	0.01
95 _Z r				180	15	4.5	19	7.6	14	5.2	12	23	0.9	27	38	20	2.2	0.34	0.16	0.33	0.67	0.49	0.27	0.18	0.02	0.01	0.01
144Ce				18			23	6.7	24	85	59	13	30	48	27	4.2	1.2	1.5	0.3	0.1	0.1	0.2	0.03	0.02	90.0	0.05	0.03
TRE(-Ce)				77	30	11	26	110	160	150	140	110	240	94	48	24	11	9.4	13	5.9	4.9	8.5	4.4	4.6	4.7	2.9	5.2
90SR				150	38	29	72	130	140	93 ,	100	83	150	09	28	22	9.4	7.8	9.9	3.4	3.0	5.1	2.8	3.1	3.9	3.4	6.5
106Ru				110	23	18	15	26	II	31	29	09	42	520	1900	2000	1400	430	191	69	29	17	Ŋ	_		0.5	_
137 _{Cs}				77	19	20	6.6	6.4	22	63	170	83	55	9/	31	15	5.6	3.5	0.9	2.1	1.6	2.7	1.1	1.4	2.0	0.9	1.7
Gross Beta	900	900 200	464	718																							
Year	1944	1946 1947	1948	1949	1950	1951	1952	1953	1954	1955	1956	1957	1958	1959	1960	1961	1962	1963	1964	1965	1966	1961	1968	1969	1970	1971	1972

a Values calculated from data supplied by Applied Health Physics Section, ORNL.

Note: Prior to 1949, analysis of water samples was limited to gross beta and gamma determinations.

b Total rare earths less cerium.

TABLE 7 $\hbox{Oak Ridge National Laboratory}$ Estimated $^{90} \hbox{Sr (In Curies*) in Process Waste Treatment Sludge Pit}$

90Sr
•
8
52
58
20
. 14
26
20
12
6
14
16
9
6
6
6
15

^{*} Estimated by assuming a $^{90}\mathrm{Sr}$ removal efficiency of 80% at the Process Waste Treatment Plant.

TABLE 8 Oak Ridge National Laboratory

Radioactive Waste (In Curies) Discharged from ORNL Stacks

Date	131 _I	Inert Gases*
1962	120	
1963	54	•
1964	84	
1965	19	
1966	16	
1967	22	
1968	10	
1969	16	76,700
1970	1.5	91,300
1971	3.5	85,500
1972	1.5	90,300

^{*}In April, 1969, the instruments used for monitoring inert gases from the 3039 and 7911 stacks were modified and calibrated to give a quantitative indication of the level of activity being released from these stacks. These figures are calculated values based on an MPC for noble gases of 2.8 x $10^{-8}~\mu\text{Ci/cm}^3$ derived for a mixture of freshly produced fission gases. These calculations assume that the noble gas is 133Xe and the values reported are "less than" values.

TABLE 9
Paducah Gaseous Diffusion Plant

Uranium Released* (Estimated Ci)

Year	Atmosphere (Offsite)	Stream (Offsite)	Soil (Onsite)
1952	< 0.01	< 0.01	< 0.01
1953	0.2	0.04	0.01
1954	1.6	< 0.01	0.08
1955	2.8	0.04	0.2
1956	3.5	< 0.01	0.3
1957	1.3	0.3	0.07
1958	1.2	0.3	0.06
1959	1.1	0.3	0.06
1960	1.0	0.6	0.1
1961	1.2	0.2	0.06
1962	0.8	0.6	0.03
1963	0.8	0.3	0.04
1964	0.3	0.3	0.02
1965		0.3	
1966	< 0.01	0.3	< 0.01
1967		0.3	
1968	0.2	0.3	0.01.
1969	0.6	0.4	0.03
1970	0.3	0.4	0.02
1971	0.4	0.4	0.02
1972	0.4	1.1	0.01

^{*} The data in this table are unofficial estimates and include measured and estimated quantities. The information was obtained from Material Release Reports, SS Material Records, and effluent monitoring data. All curie quantities of uranium are based on normal ²³⁸U activity (3000 kg/curie). Most of the uranium was normal or depleted in the ²³⁵U isotope.

TABLE 10

Paducah Gaseous Diffusion Plant

Uranium Discards to Holding Pond and Burial Areas (Onsite)

Area Description	Period of Use	Primary Type of Scrap	Approx. Quantity Curies l
C-749 Uranium Burial Ground	1957-1972	Depleted Uranium Metal	80
C-404 Holding Pond - Changed to Open Pit for Solid Waste in 1957	1953-1957	Uranium Liquid Waste from C-400	1
	1957-1972	Metals Plant Slag, Rejected Depleted UF ₄ , Filter Cake	573

 $^{^{1}}$ All curie quantities of uranium based on normal ^{238}U activity (3000 kg/curie).

TABLE 11
Paducah Gaseous Diffusion Plant

²³⁰Th Released¹ 1956-1972

Released To	Estimated Quantity Curies
C-404 Holding Pond and Solid Waste (Onsite)	0.1
Stream (Offsite)	0.1

 $^{^{\}rm l}$ These determinations were based on known presence in UF $_{\rm 6}$ cylinder wash solutions and from uranium and $^{\rm 237}{\rm Np}$ recovery waste solutions. The material was released in low concentrations.

TABLE 12
Oak Ridge Y-12 Plant

Gaseous and Liquid Waste Stream (Estimated Ci) $^{\rm l}$

Fiscal	Su	rface Streams			Stacks ²	
<u>Year</u>	Enriched U	Depleted U	Thorium	Enriched U	Depleted U	Thorium
1954	_	.01	< .01	_	•	
1955	-	.02	< .01	_		
1956	_	.3	< .01	0.2		
1957	-	.6	< .01	0.2		
1958	-	.3	< .01	0.3		
1959	-	• 4	.34	0.4		
1960	2.8	.3	.03	0.5		
1961	0.6	.1	.09	0.4		
1962	0.7	.02	-	0.6		
1963	0.7	.01	< .01	0.8		
1964	0.7	.01	< .01	0.4		
1965	1.1	< .01	-	0.4		
1966	0.8	.02	-	0.5		
1967	0.7	.01	-	0.4		
1968	0.8	.01	-	0.3		
1969	0.8	.01	-	0.4	.03	< .01
1970	0.6	.02		0.4	.02	< .01
1971	0.8	.01	-	0.4	< .01	< .01
1972	0.8	.8		0.6	< .01	< .01

The curie quantities were estimated from the information obtained from the Y-12 Plant's Nuclear Material Inventory Report.

² Information not available for stack releases for depleted uranium and thorium prior to 1969.

TABLE 13

Oak Ridge Gaseous Diffusion Plant

Uranium Released¹ (Estimated Ci)

Year	Atmosphere (Offsite)	Ground (Onsite) ²	Stream (Offsite) ³
1946 ⁴		2.0	•
1947		0.05	
1948	0.01	0.01	
1949		0.03	
1950		< 0.01	
1951	•		•
1952	0.3	< 0.01	
1953	0.3	0.09	
1954	0.1	0.02	
1955	0.1	0.03	
1956	0.3	0.03	
1957	0.2	0.02	
1958	0.2	0.03	
1959	0.3		
1960	1.0	0.01	
1961	1.0	0.1	
1962	0.02	0.3	
1963 ⁵	0.1		
1964	0.01		
1965	0.01	< 0.01	
1966		0.01	
1967		0.04	
1968		< 0.01	
1969	0.1	•	0.3
1970	0.1	0.1	0.4
1971	0.3	0.02	0.3
1972	0.1		0.4

Footnotes to Table 13

- The data in this table are unofficial estimates and include measured and estimated quantities. All curie quantities are based on the equation: $dis/min/\mu g = 0.75 + 1.06$ (wt % ²³⁵U).
- Portions of the material released to the ground eventually would have been received by an offsite water stream.
- The estimated quantity released to an offsite water stream was based on effluent monitoring. This type sampling program was not in effect prior to 1969. Neither is analytical information available for this period.
- 4 Little analytical information is available for the earlier operating years of the plant.
- Analytical and data records were not maintained fully from 1963 to 1968 due to a reduction in the work force at the ORGDP.

TABLE 14 Oak Ridge Gaseous Diffusion Plant Uranium Disposed Of In Solid Form¹

Location	Activity <u>(Curies)</u>
K-1407-B Holding Pond	7.3
ORGDP Burial Ground	11.3
ORNL Burial Ground	2.0
Sludge Disposal Area	0.2
Y-12 Burial Ground	0.9

The data in this table are unofficial estimates and include measured and estimated quantities. All curie quantities of uranium are based on: $dis/min/\mu g = 0.75 + 1.06$ (wt % $^{2.35}U$).

REFERENCE MATERIALS

- 1. ORNL Radioactive Waste Management Plans, ORO-723 (ORNL), June 1973.
- 2. The Occurrence and Retention of Radionuclides in the Sediments of White Oak Lake, T. F. Lomenick and D. A. Gardiner, Health Physics, Vol. II No. 6, June 1965.
- 3. Environmental Monitoring Summary for the Paducah Plant, KY-273, 1958.
- 4. Environmental Monitoring Summary for the Paducah Plant, KY-332, 1959.
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- 6. Environmental Monitoring Summary for the Paducah Plant, KY-419, 1961.
- 7. Environmental Monitoring Summary for the Paducah Plant, KY-458, 1962-1963.
- 8. Environmental Monitoring Summary for the Paducah Plant, KY-484, 1964.
- 9. Environmental Monitoring Summary for the Paducah Plant, KY-543, 1965-1966.
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- 11. Environmental Monitoring Summary for the Paducah Plant, KY-624, 1969.
- 12. Environmental Monitoring Summary for the Paducah Plant, KY-629, 1970.
- 13. Environmental Monitoring Report, United States Atomic Energy Commission, Paducah Gaseous Diffusion Plant, UCC-ND-222, 1971.
- 14. Environmental Monitoring Report, United States Atomic Energy Commission, Paducah Gaseous Diffusion Plant, UCC-ND-245, 1972.
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- 17. AEC Radioactive Effluent Reports, 1971.
- 18. AEC Radioactive Effluent Reports, 1972.
- 19. Radioactive Waste Management Plans Paducah Gaseous Diffusion Plant, UCC-ND-68, January 1969.
- 20. Radioactive Waste Management Plans Paducah Gaseous Diffusion Plant, UCC-ND-214, August 1972.
- 21. Radioactive Waste Management Plans, ORO-723 (PAD), June 1973.
- 22. Nuclear Materials Inventory Report Y-12 Plant.
- 23. Radioactive Waste Management Plans, ORO-723 (ORGDP), June 1973.
- 24. FZA (AEC), Uranium Control Department, SS Material Control Report, K-P-3530, Part 6, December 1972.
- 25. Oak Ridge Gaseous Diffusion Plant Quarterly Report, Second Quarter FY 1969, K-1806, October 1, 1968 through December 31, 1968.
- 26. Uranium Hexafluoride Handling Procedures and Container Criteria, ORO-651 Revision 3, August 1972.

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